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Improvement of soft magnetic properties by simultaneous addition of P and Cu for nanocrystalline FeNbB alloys

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The additional effect of P and/or Cu on the structure and the magnetic properties of Fe–Nb–B nanocrystalline soft magnetic alloy tapes with a wide width of 5 mm produced in air was investigated. Although Cu or P addition changes the magnetic softness a little, only the simultaneous addition of 1 at. % P and 0.1 at. % Cu significantly improves the soft magnetic properties of the crystallized alloys. The best magnetic properties (higher permeability than 45×10^3, lower coercivity than 5 A/m, and saturation magnetic flux density of 1.50–1.54 T) were obtained in the compositional ranges of 6.6–6.8 at. % Nb and 8.4–8.8 at. % B with 1% P and 0.1% Cu, and these values are superior to the typical nanocrystallized $Fe_{84}Nb_7B_9$ alloy prepared in a vacuum. The improvement probably originates from the decrease in the distribution of the α-Fe grain size in the crystallized structure by the simultaneous addition. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2714676]

I. INTRODUCTION

It has been already reported that the nanocrystalline $Fe_{84-90}M_7B_{3-9}$ (M=Zr,Hf,Nb) alloy "nanoperm" exhibits high saturation magnetic induction (B_s) of 1.5–1.7 T as well as good soft magnetic properties.[1–4] The crystallized Fe–M–B alloys consist of α-Fe nanocrystallites about 10–15 nm in size embedded in an amorphous minority matrix. However, the melt spinning in air is difficult, which is a disadvantage for mass production, because the M elements are easy to be oxidized. We have already reported that the nanocrystalline $Fe_{83.9-85.9}Nb_{5.0-6.0}B_{8.0-9.0}P_{1.0}Cu_{0.1}$ alloys can be produced in air and exhibit higher B_s than 1.5 T and good soft magnetic properties, simultaneously.[2,3] The best magnetic properties, i.e., B_s of 1.61 T and permeability (μ) of 41×10^3 at 1 kHz, were achieved in $Fe_{84.9}Nb_{6.0}B_{8.0}P_{1.0}Cu_{0.1}$. The only simultaneous addition of 1 at. % P and 0.1 at. % Cu drastically changes the as-quenched structure from an amorphous phase including a tiny fraction of the α-Fe grain about 40–50 nm in size to an amorphous phase with some other phase, an amorphous or α-Fe like cluster, less than 2 nm in size, which probably results in decrease in the α-Fe grain size dispersion at the crystallized state. However, the previous results were obtained for the ribbon specimens with a limited alloy composition of 6 at. % Nb and the very narrow width of approximately 1 mm. It is necessary to examine the soft magnetic properties of the wide melt-spun tapes for practical application. In general, the nanocrystallized structure strongly depends on the precursor, as-quenched structure, which should change with the quenching rate, and the quenching rate should decrease with the ribbon width. In this study, the additional effect of P and/or Cu on the soft magnetic properties and the structure was investigated in detail for Fe–Nb–B alloy ribbons with a relatively wide width of 5 mm produced in air.

II. EXPERIMENTAL PROCEDURE

Alloy ingots were prepared by arc-melting a mixture of pure Fe (3N), Nb (3N), and Cu (4N) metals, premelted Fe–P, and pure B (2N5) crystal in an Ar atmosphere. A single-roller melt-spinning method in air was used to produce the rapidly solidified tapes about 5 mm in width and approximately 20 μm in thickness. The alloy compositions are nominally expressed since the difference between nominal and chemical analyzed composition was negligibly small. The as-quenched tapes were wounded into a toroidal shape to be used as specimens. Crystallizing treatment was carried out by treating the specimens for 300 s at various temperatures in a vacuum with a heating rate of 3 K/s. The as-quenched and the annealed structures were identified by x-ray diffractometry (XRD). The mean grain size of α-Fe was estimated using Scherrer's equation from the full width at half maximum of the (100) x-ray reflection peak. The permeability (μ) was measured by a vector impedance analyzer at 1 kHz under a field of 0.4 A/m. The saturation magnetic flux density (B_s) and the coercivity (H_c) under a maximum applied field

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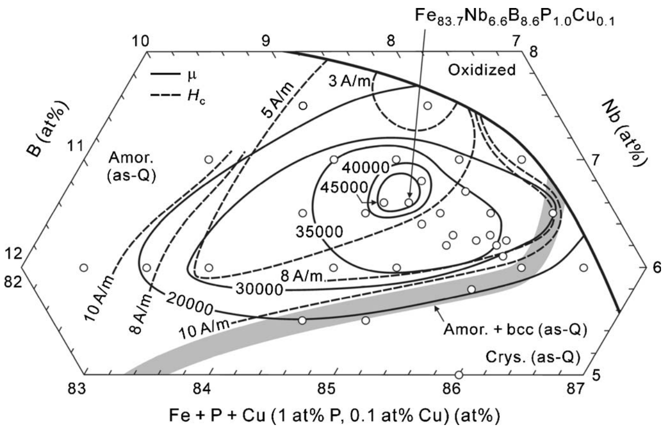

FIG. 1. Compositional dependence of permeability at 1 kHz (μ) and coercivity (H_c) of crystallized $FeNbBP_{1.0}Cu_{0.1}$, along with the phase field in an as-quenched state.

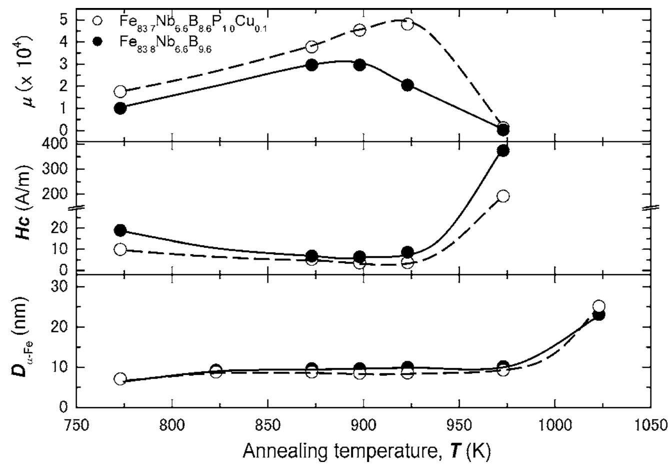

FIG. 2. Dependence of mean grain size of α-Fe ($D_{\alpha\text{-Fe}}$), coercivity (H_c), and permeability at 1 kHz (μ) on annealing temperature for $Fe_{83.8}Nb_{6.6}B_{9.6}$ (black circle) and $Fe_{83.7}Nb_{6.6}B_{8.6}P_{1.0}Cu_{0.1}$ (open circle) alloys.

of 800 kA/m were measured by a vibrating sample magnetometer and a dc *B*-*H* loop tracer. All the measurements are carried out at room temperature.

III. RESULTS AND DISCUSSION

The pseudoternary diagram of the magnetic properties of the nanocrystallized Fe–Nb–B–$P_{1.0}Cu_{0.1}$ alloy tapes annealed at 923 K for 300 s with 5 mm width is shown in Fig. 1. The as-quenched structure of the tapes identified by XRD is also shown in Fig. 1. Hence, the change of the amorphous-forming ability is considered to be little by the substitution of P and Cu for B and Fe, respectively, because Cu has no amorphous-forming ability and P has slightly lower ability than B for the Fe based alloys. The thick line shows the limit of the melt spinning in air; the remarkably oxidized tapes were obtained in the upper region of the thick line. The higher μ than 40×10^3 was obtained in the compositional ranges of 6.6–6.8 at. % Nb and 8.4–8.8 at. % B. The highest μ is 48×10^3 for $Fe_{83.7}Nb_{6.6}B_{8.6}P_{1.0}Cu_{0.1}$: this is superior to the reported value (36×10^3) for $Fe_{84}Nb_7B_9$ alloy.[1]

Figure 2 shows the annealing temperature dependence of the α-Fe grain size, μ, and H_c for $Fe_{83.8}Nb_{6.6}B_{9.6}$ and $Fe_{83.7}Nb_{6.6}B_{8.6}P_{1.0}Cu_{0.1}$ alloys with the best soft magnetic properties. Both as-quenched alloys were confirmed to be amorphous by XRD and show the crystallization by the annealing. The best soft magnetic properties were obtained in the temperature range from 898 to 923 K for P and Cu added alloy and from 873 to 898 K for the ternary alloy. The mean grain size of α-Fe is slightly smaller except at 1023 K and the soft magnetic properties, μ, and H_c are superior for the P and Cu added alloy in whole annealing temperature range. The α-Fe grain size distribution of the P and Cu added and ternary alloys determined by transmission electron microscopy is shown in Fig. 3. The distribution of the P and Cu added alloy is much narrower than that of the ternary alloy; the standard deviation significantly deceases from 0.391 to 0.236 by the addition. We have already reported that the soft magnetic properties strongly depend on the grain size and the dispersion of the α-Fe grains and the volume fraction of the α-Fe phase for the nanocrystalline soft magnetic Fe–Nb–B alloys.[5] The volume fraction of the α-Fe phase evaluated by the XRD profiles was about 37% for both alloys. Therefore,

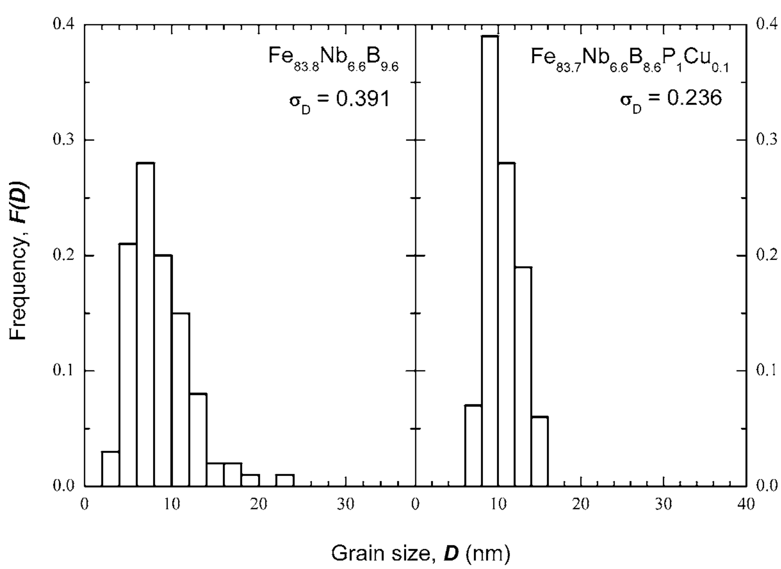

FIG. 3. α-Fe grain size distribution for $Fe_{83.8}Nb_{6.6}B_{9.6}$ and $Fe_{83.7}Nb_{6.6}B_{8.6}P_{1.0}Cu_{0.1}$ alloys annealed at 923 K.

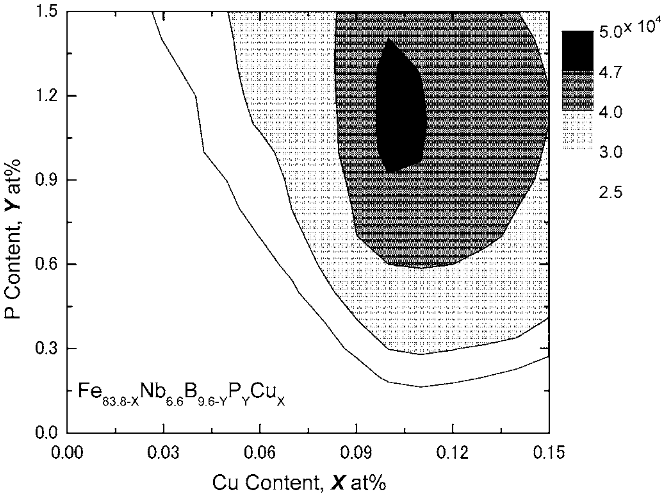

FIG. 4. Dependence of permeability at 1 kHz on P and Cu contents for $Fe_{83.8-X}Nb_{6.6}B_{9.6-Y}P_YCu_X$ alloy annealed at 923 K.

the narrow distribution of α-Fe grain size should be a dominant reason for the improvement of the soft magnetic properties by the P and Cu addition.

Figure 4 shows the change μ after annealing at 923 K for 300 s as a function of P and Cu contents for $Fe_{83.8-X}Nb_{6.6}B_{9.6-Y}P_YCu_X$. The maximum value is obtained for the simultaneous addition of 1 at. % P and 0.1 at. % Cu and gradually decreases with deviation from the alloy compositions. The same effect of the addition has been obtained for a slightly different alloy composition, $Fe_{84.9-X}Nb_{6.0}B_{9.0-Y}P_YCu_X$.[2,3]

Figure 5 shows the mixing enthalpy between the constituent elements.[6] The value between Fe and Cu is positive,

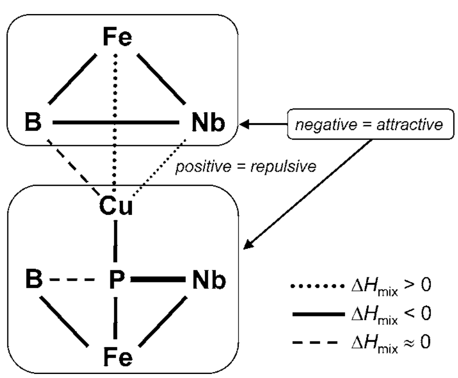

FIG. 5. Mixing enthalpies between constituent elements in the Fe–Nb–B–P–Cu alloy system.

therefore, a repulsive interaction should exist between Fe and Cu atoms. On the other hand, an attractive interaction should exist between Cu and P, P and Fe, and P and Nb. Therefore, the as-quenched structure possibly consists of two phases; the major part is an Fe–Nb–B based amorphous phase, and the minor part is a Cu containing phase. We have reported that the as-quenched structure of $Fe_{84.9}Nb_6B_8P_1Cu_{0.1}$ alloy is revealed by transmission microscopy to be composed of an amorphous phase as a major part and a large number of extremely small (less than 2 nm) grains with amorphous phase or α-Fe-like cluster as a minor part.[2] The minor Cu containing phase, if the phase is amorphous, should have lower thermal stability than the major amorphous phase, because the Cu addition to Fe based amorphous alloys is well known to decrease the crystallization temperature. If the minor phase is an α-Fe-like cluster, it should be the nucleation site of α-Fe phase. Therefore, the phase probably acts as the nucleation site for α-Fe and which presumably results in the decrease in the α-Fe grain size and the improvement of the soft magnetic properties for the nanocrystalline alloys.

IV. CONCLUSIONS

We have investigated the additional effect of P and Cu on the structure and soft magnetic properties for Fe–Nb–B alloys with 5 mm width melt spun in air. The obtained results are summarized as follows. The simultaneous addition of 1 at. % P and 0.1 at. % Cu significantly improves the magnetic softness of the crystallized $Fe_{83.6}Nb_{6.6}B_{9.6}$ alloy; μ increases from 29 000 to 48 000 and H_c decreases from 6.88 to 3.74 A/m after the optimum annealing, probably resulting from the narrowed distribution of α-Fe grain size by the simultaneous addition of 1 at. % P and 0.1 at. % Cu.

The best magnetic properties ($\mu > 45 \times 10^3$, $H_c < 5$ A/m and $B_s = 1.50-1.54$ T) were obtained for the compositional ranges of 6.6–6.8 at. % Nb and 8.4–8.8 at. % B with 1 at. % P and 0.1 at. % Cu. These values are superior to those for the typical $Fe_{84}Nb_7B_9$ nanocrystalline alloy.

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